Transport Pathways of Fire Generated Tracers to the Upper

Troposphere over South America and Africa as determined by

the A-Train satellite measurements

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Abstract

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We use carbon monoxide (CO) measurements from MLS and TES, and cloud water content measurements from CloudSat to identify the transport pathways of pollutants generated by tropical biomass burning to the upper troposphere (UT). There appeared to be three pathways that transport CO from its surface sources to the UT: "local convection", "UT advection" and "lower troposphere (LT) advection → convection". The dominant transport pathway varies geographically. The "local-convection" and "LT advection → convection" pathways were the main contributors for high CO in the UT over Africa, whereas long-range "UT advection" pathway appeared to transport CO to the UT over tropical South America and Atlantic Ocean. This case study suggests that our method of identifying these pathways through joint use of multiple A-Train satellite sensors can potentially be applied to address the need of characterizing and understanding the climatological variations of the dominant pathways for CO transport to the UT.

1 Introduction

Biomass burning has a significant influence on the chemical composition of the free troposphere and has been recognized as a globally important source of many trace gases and aerosols. Among them, carbon monoxide (CO) plays an important role in atmospheric chemistry and radiation balance. With a lifetime of 1-2 months in the troposphere, CO is an excellent tracer of the transport of polluted air masses. Identifying the distribution of CO concentration at different altitudes is important to the understanding of the mass transport for biomass and fossil fuel burning.

Africa and South America are two major source regions for tropical biomass burning owing to forest clearance, savanna burning, and land management. African biomass burning occurs during late November to early March in the Northern Hemisphere (NH) and July to October in the Southern Hemisphere (SH) [Giglio et al., 2006], whereas South American biomass burning peaks in August - October. Land convection has an important impact on CO transport from surface sources to the lower stratosphere in the tropics and Africa emerges as a major convective transport center [Ricaud et al., 2007; Liu et al., 2007]. Although biomass burning has been hypothesized to be a potential source of the semi-annual peaks of CO in the upper troposphere (UT) and lower stratosphere [Schoeberl et al., 2006], it is still debatable as to how CO emitted over these two tropical continents contributes to the CO centers in the UT.

The large spatial extent of biomass burning over South America and Africa and sparseness of the ground based observations make remote sensing the only feasible way of detecting and monitoring their related transport events. In recent years, CO observations from airborne and spaceborne instruments have been extensively used to study the inter-continental transport of pollutants in the troposphere [e.g. *Duncan et*

al., 2003]. Since August 2004, the Aura Microwave Limb Sounder (MLS) and Tropospheric Emission Spectrometer (TES) have provided unprecedented information about the vertical distributions of CO globally [Rinsland et al., 2006; Livesey et al., 2008]. Previous studies using data from these instruments have demonstrated the utility of the A-Train satellites in identifying and understanding the long-range CO transport pathways from its surface sources to the UT and lower stratosphere [e.g. Fu et al., 2006; Jiang et al., 2007]. However, a systematic survey of the characteristics of the CO transport pathways, especially the relative importance of different pathways on climate scales, is still not available. As a first step to address this problem, we aim to develop a methodology to automate identification of these pathways using multiple A-Train satellite sensors and to explore types of transport pathway that control CO concentration and variation in the UT over the two continents through a case study.

2 Data and Methodology

We use fire count data from Aqua Moderate-resolution Imaging Spectroradiometer (MODIS) to locate biomass burning events, Aura TES and MLS CO data to determine changes of CO profiles in the lower and upper troposphere, and CloudSat cloud water content (CWC) data, which is the combination of liquid/ice water content (L/IWC), to determine the occurrence and structure of convection.

The MODIS fire data is the total number of fire events counted for each 0.5° grid and 8-day period globally. The footprint of each TES nadir observation is 5×8 km, averaged over detectors [*Beer*, 2006]. Aura TES CO retrieval is most sensitive to CO concentration between 700 and 200 hPa and the uncertainty is about 10-20% [*Luo et*

- al., 2007]. MLS CO data has vertical resolution ~4 km, and horizontal resolution 68 ~300 km. At 147 hPa, the uncertainty of MLS CO measurements is about 30%. At 69 70 215 hPa, the current MLS retrieval overestimates CO by about a factor of 2, although this bias does not alter the spatial pattern of the UT CO [Livesey et al., 2008]. 71 72 CloudSat provides CWC measurements in less than 6 minutes before MLS and 14 minutes before TES [Stephens et al., 2002]. The vertical resolution of CloudSat data is 73 ~500m. The horizontal resolution is ~2 km. To homogenize the horizontal resolutions 74 75 of the MLS, TES and CloudSat data and also match the temporal resolution of the 76 MODIS fire data, we averaged the MLS, TES and CloudSat data onto 4° latitude $\times 8^{\circ}$ longitude grid boxes and over 8-day period. We also use three dimensional wind 77 78 velocity data between 1000 and 100 hPa from the NCEP reanalysis for the same time 79 period as the MLS observations. The daily wind data are also averaged over the same 8-day period with the horizontal resolution of $2.5^{\circ} \times 2.5^{\circ}$. 80
- 81 Previous observations, primarily from field campaigns, have suggested that fire 82 generated tracers could be transported to the UT by three pathways:
- 83 a. Local convection, including pyro-convection, transports a tracer directly from the surface to the UT [e.g. Pickering et al., 1996]. We refer to this pathway as 84 the "local-convection" pathway;

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b. Advection in the lower troposphere (LT) transports a tracer from its source region to the tropical convective region, and then convection lifts the tracer to the UT [Andreae et al., 2001]. We refer to this pathway as the "LT advection→ convection" pathway.

c. Convection in remote source region transports a tracer to the UT, then advection carries the tracer downstream in the UT [Ray et al., 2004]. We refer to this pathway as the "UT advection" pathway;

To identify these transport pathways using satellite measurements, we define (a) The "local-convection" pathway: when fires, deep convection and increase of CO in the UT are simultaneously detected and co-located as shown by MODIS, CloudSat, TES and MLS; (b) The "LT advection→ convection" pathway: when convection and increase of CO in the UT occur simultaneously without fires; (c) The "UT advection" pathway: when high CO concentrations are detected in the UT in absence of convection and fires at the same time and location.

3 Results and Discussions

The domain we choose is $50^{\circ}\text{S} \sim 50^{\circ}\text{N}$ latitude, $100^{\circ}\text{W} \sim 100^{\circ}\text{E}$ longitude (Figure 1). During the period August 5-12 of 2007, numerous fire events occurred over the South African and South American continents. Figure 1 shows distributions of fire counts and convective clouds during this period in our domain, and the vertical cross sections we chose for the analysis. The major centers of fires were all located in the SH, although secondary centers of fires also occurred in southeastern US and northern Europe. As shown by the CloudSat cloud water path data, nearly all the fire events were located in the dry regions where convection is very weak. The fire events in South Africa were much more frequent than those in South America and most intense in south Congo, Angola and Zambia. In South America, most fire events happened in the southeastern Amazon. In order to compare the distributions of CO and convection

at different locations including land and ocean surface, we chose three meridional cross sections located at 60°W, 20°W and 12°E (Figure 1 and 3), which pass through South America, the Atlantic Ocean and Africa respectively. We also chose three zonal cross sections in the tropical SH (10°S), NH (18°N) and near the equator (2°N).

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Figure 2 shows the CO distribution in the lower and middle troposphere (Figure 2a-2b) as measured by TES and that in the UT (Figure 2c-2d) as measured by MLS for the period August 5-12 of 2007. Centers of high CO in the lower and middle troposphere over Africa in the SH mostly overlapped with those of fires (Figure 2a-2b). By contrast, high CO in the lower and middle troposphere over South America was located over the Andes to the west of the fires in the Amazon. These different relative locations between lower to middle tropospheric CO and fires at the surface are presumably caused by differences in daytime boundary layer depth and circulation between the two tropical continents. The daytime boundary layer of drier savanna regions over Africa is expected to be much higher than that over the humid Amazon rainforests, and could transport CO to the middle troposphere. Thus, CO could be lifted to lower and middle troposphere by strong boundary layer turbulence within the fire regions. By contract, the boundary layer over the humid Amazon is usually shallower than 1.5 km. CO has to be advected to the eastern slope of Andes to reach 700 hPa or higher altitude in order to be detected by TES. Thus, how CO generated at the surface is ventilated out of the atmospheric boundary layer depends on the depth of the atmospheric boundary layer and circulation pattern. In the UT, the centers of high CO concentration were displaced further away from the fire regions. While most

of the fires occurred south of the equator (Figure 1), the centers of highest CO concentration were located north of the equator, either within convective areas or the downwind areas of convection (Figure 2c-2d). This is not surprise because agricultural burning in forests and savanna regions tends to occur during dry season when deep convection is infrequent and large-scale subsidence prevails. Figure 2 highlights the importance of long-range advection in the LT and UT.

To determine transport pathways from the surface to the UT, we use Figure 3 to examine the vertical distribution of CO and CWC along several meridional and zonal cross sections as marked in Figure 1. Large values of CloudSat CWC are indication of deep convective clouds. The MODIS fire counts are scaled in order to show the spatial distribution and relative number of fire events in each cross section. Along the 60°W cross section over the American continents (Figure 3a), the fire events and high CO in the LT were concentrated at 25°-5°S region, above which subsidence dominated. There was no convection directly above the fire region, instead strong convection occurred on the north and south sides adjacent to the burning area.

In the UT, CO was highest to the north of the equator (5°-40°N), where deep tropical convection occurred. However, the weak northward advection in the LT and low CO concentration in the middle troposphere of the convective areas (0°-30°N) suggest that the high CO in the UT over South America was probably not transported by local convection. This contention is supported by Figure 2a which shows that the lower tropospheric air over the northern South America to the north of the fire regions is mainly advected from the Atlantic Ocean with low CO concentration. How was CO

transported to the UT over the subtropical South America? The northeasterly winds in the UT (Figure 2c-2d) suggest that the air with high CO over the northern South America was likely transported from the southeast coast of US. Thus, high CO in the UT over the tropical South America was transported via a "UT advection" pathway.

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Over the African continent (Figure 3b), large number of fire events peaked around 10°S, in part overlap with the areas of deep convective clouds and strong rising motion (10°S-10°N). High CO in the middle troposphere peaked at 5°S, on the north side of the fire center. The northward displacement of middle tropospheric CO relative to its surface source region is probably due to both local convective transport and LT advection that occurred over the northern end of the fire regions (Figure 3b and 2a). Thus, both "local-convection" and "LT advection→ convection" pathways were contributors to the high CO center in the UT over the equatorial western Africa. To the north of the equator, there was a broad area with relatively higher CO in the UT. Given lack of fire at the surface, lack of high CO in the middle troposphere, and prevailing easterly wind in the UT, the CO is likely advected from the Indian subcontinent where a large high CO center was located (Figure 2c-2d), via a "convection → UT advection" pathway. Thus, CO in the UT over the tropical Africa was transported by "local-convection", "LT advection→ convection" and "UT advection" pathways from local and remote biomass burning areas.

High CO concentration also appeared over much of the tropical Atlantic Ocean.

To the north of the equator, the high CO appears to be mainly caused by easterly advection in the UT from Africa (Figure 2c-2d). To the south of the equator, relatively

high CO appears in the lower, middle and upper troposphere (Figure 2). Figure 3c shows a higher CO center below 5 km above the sea level ranged from 5°N-15°S. Given the strong subsidence throughout the troposphere over the southern tropical Atlantic Ocean and low CO between 5 km and 10 km, the higher CO in the UT over this region must be advected by northeasterly wind from the equatorial western Africa (Figure 2c). Thus, the "UT advection" pathway is likely responsible for the observed high CO in the UT over the tropical Atlantic Ocean.

The zonal long-range transport is investigated using three zonal-vertical cross sections as shown in Figure 3d-3f. At 10°S (Figure 3d), MODIS and TES showed much stronger fire events and higher CO in the lower to middle troposphere over Africa (15°-40°E) than that over South America (40°-70°W). By contrast, the CO concentration in the UT was comparable between the two continents. CloudSat showed that the convection over the Andes only reached about 6 km above the sea level, thus unable to transport lower tropospheric CO to the UT. CO in the UT was more likely advected from North America (Figure 2c-2d). Over the Atlantic Ocean and western Africa sector (20°E-30°W), deep convection was also absent. High CO air was advected by northeasterly wind in the UT from western Africa in the NH. Figure 2c, 2d and 3d together suggest that the "UT advection" is the main transport pathway for UT CO in this sector.

Near the equator (2°N, Figure 3e), strong convective systems developed above both South America and Africa. However, very few fire events occurred on the continental surface. The strong convection penetrated to about 15km altitude and CO

concentration was high in tropospheric column over the convective region in Africa. High CO in the LT over Africa as shown by TES was advected from the southern fire region. This combination of strong convection near the equator and advection of CO from the fire region in the SH suggests that the "LT advection -> convection" pathway was responsible for high CO in the UT over Africa. Although higher CO was also shown in the LT over the Atlantic Ocean, due to lack of convection and relatively low CO in the middle troposphere, the high CO in the UT over the Atlantic Ocean was mostly advected from Africa, via a "UT advection" pathway. Over South America, despite of present of strong convection, few fire events and low CO in the lower and middle troposphere suggest that the "local-convection" pathway is not the main transport mechanism. Instead, the "UT advection" of CO from SE US and the Gulf of Mexico was likely responsible for the UT CO over the equatorial South America. In the NH (18°N, Figure 3f), fire events were absent except for very few in south Mexico (80°-100°W). CO concentration was low in the lower and middle troposphere across the entire Africa-Atlantic-America sector. Thus, the high CO concentration in the UT must be transported by the "UT advection" from the SE US, the equatorial Africa and the Indian subcontinent. Figure 3 suggests that "local-convection", "LT advection

convection" and "UT advection" pathways all contributed to the UT CO distribution over the tropical Africa, whereas the "UT advection" pathway dominated the UT CO over South America and the Atlantic Ocean. We have also analyzed other

4 Conclusions

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cross sections and found similar pathways as shown in Figure 3.

We have tested the joint use of A-Train satellite measurements for automating identification of the pathways for transporting fire produced CO into the UT for the period of August 5-12, 2007. Aura MLS and TES are used to determine the distribution of CO in the UT, LT and middle troposphere, respectively. The CloudSat CWC is used to identify convective clouds and the Aqua MODIS fire count product is used to identify the biomass burning fire events. Over the African continent, CO is transported by the "local-convection" and "LT advection → convection" pathways from the fire regions of Africa in the SH to the UT over the tropical Africa, and the "UT advection" pathway contributed to the long-range transport of CO from the remote CO source region over the Indian subcontinent to the UT over Africa. Transport by the "local convection" mainly occurred in the areas where the margins of convective areas and fire areas overlap with each other. Over South America, the local generated CO does not appear to be the main contributor to the UT CO. Instead, the "UT advection" from North America, especially from the SE US may be the main source of high UT CO over the tropical South America. The "UT advection" pathway also appeared to dominate the UT CO over the Atlantic Ocean. Our results reveal that the dominant transport pathways for the UT CO vary

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Our results reveal that the dominant transport pathways for the UT CO vary geographically, presumably depending on the surface climate conditions and the circulation patterns. Thus, an observational characterization of the "climatology" and variations of these transport pathways are needed in order to adequately understand the distribution of the biomass burning generated pollutants in the tropics. The method we developed in this study could be applied to longer period to assess the

- 244 climatological distribution of the transport pathways over the tropical continents, and
- their relative importance.
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Figure Captions

- Figure 1. Distribution of the fire events during the period of August 5-12, 2007 over
- 298 Africa and South America. The white-blue shaded color indicates the distribution of
- 299 CloudSat cloud water path (g/m²). The locations of cross sections are indicated by
- 300 black bold straight lines.
- Figure 2. Mean concentrations of CO at different pressure levels during the period of
- August 5-12, 2007 obtained from TES (a-b) and MLS (c-d) observations respectively,
- over South American and African continents. The MLS CO at 215 hPa is divided by
- 304 1.5 in order to partly offset the bias. The black vectors indicate horizontal winds
- 305 derived from the NCEP reanalysis.
- Figure 3. Distribution of MLS and TES derived CO volume mixing ratio (color-line
- 307 contour) and CloudSat cloud water content (color-shades) in the vertical-meridional
- 308 cross sections (left) and the vertical-zonal cross sections (right). The black arrows
- 309 indicate meridional and vertical winds derived from the NCEP reanalysis. Vertical

- velocity is enlarged 100 times for clarity. The thin black horizontal line indicates the
- boundary between MLS and TES CO. The dark red bold line at the bottom indicates
- the scaled fire counts at the surface of each cross section (unit: number of fire pixels).

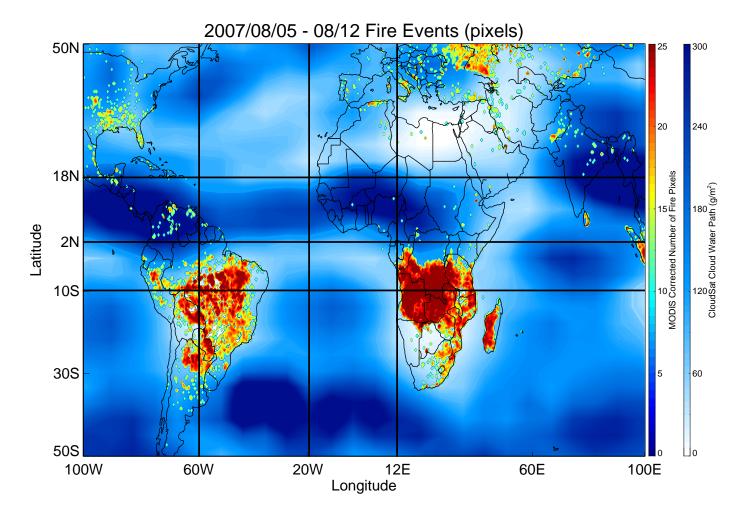


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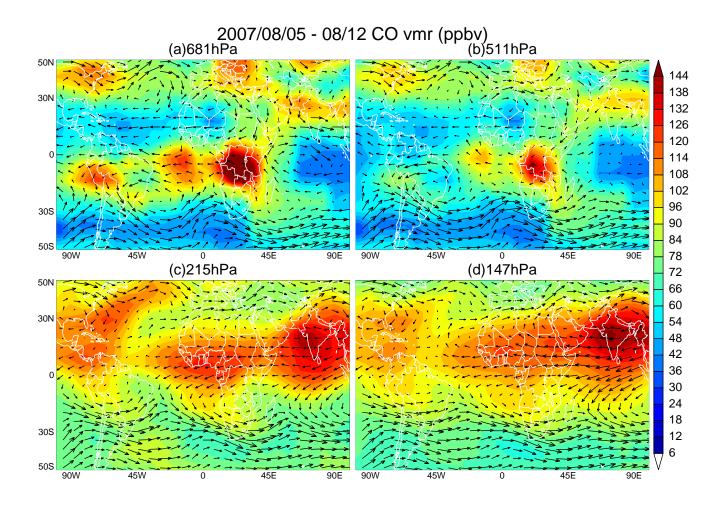


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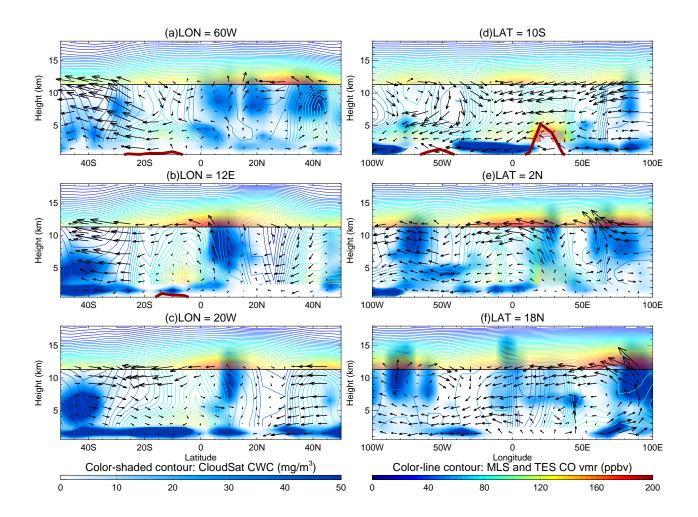


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